Discrete Change of Spin-Density-Wave Modulation in Cr(100)/Sn Multilayers as a Function of Cr Layer Thickness

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Magnetic structures of epitaxial Cr(001)/Sn multilayers, where monatomic Sn layers are periodically embedded in a Cr(001) film, were studied using neutron diffraction and Mössbauer spectroscopy. It was found that spin-density-wave (SDW) antiferromagnetic structures with the modulation along the perpendicular direction to the film plane are stabilized at low temperatures. The wavelength of the SDW discretely changes as a function of the superlattice period. The enhanced magnetic moments of Cr at the Cr/Sn interfaces are thought to pin the antimode of the SDW modulation at the interface, resulting in the SDW with the modulation harmonic to the superlattice period.

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The magnetic properties of metallic Cr are sensitively influenced by environmental conditions, such as impurities, strains, and interface effects [1]. Pure bulk Cr has a spin-density-wave (SDW) antiferromagnetic structure below the Néel temperature (311 K). For thin Cr films or Cr layers in multilayered structures, the magnetism, including the SDW state, is expected to be different from that in bulk. Most of the recent works in this field are focused on Fe/Cr layered structures, although the magnetism of the Cr layers, particularly in the thickness region where the oscillatory coupling between the Fe layers appears, is still open to discussion [2–4]. In this Letter, it is demonstrated that the spin structure of Cr can be controlled by pinning the antinode of the SDW modulation using monatomic layers of a nonmagnetic element. The system introduced here is epitaxial Cr(001)/Sn multilayers, where monatomic Sn layers (~ 2 Å) are periodically inserted in a Cr(001) film. Information on the periodic magnetic structure is obtained from neutron diffraction measurements. The local magnetic properties around the Sn layers, on the other hand, can be examined by measuring the magnetic hyperfine field induced at the Sn sites using ^{119}Sn Mössbauer spectroscopy. The magnetic structures were clarified for the multilayers with different Cr layer thickness (t_{Cr}) using the results obtained from both experimental methods.

The $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})] \times n$ multilayers were grown on MgO(001) substrates at 200 °C using an ultrahigh-vacuum deposition system with electron-gun heating. The growth conditions are the same as those for the previously reported Cr/Sn multilayers [5]. The Sn atoms are located at the substitutional sites of bcc Cr(001) planes, forming a body-centered tetragonal lattice together with the neighboring Cr atoms. Neutron diffraction measurements were performed with conventional triple-axis spectrometers, TOPAN and TAS-1, at JRR-3M in JAERI. The scans were carried out around the Cr(001) and (010) reciprocal points along the [001] axis (the L direction) and the [010] axis (the K direction). Nuclear reflections are forbidden around these reciprocal points, so that only magnetic peaks due to antiferromagnetic structures are expected to appear (for details, see Ref. [6]). Mössbauer spectra were measured by means of conversion electron Mössbauer spectroscopy, using a gas-flow counter with He + 1%$(CH_3)_3CH$ at room temperature and a gas-filled counter with H_2 at 20 K [7]. The size of the magnetic hyperfine field induced at the Sn sites is thought to be roughly proportional to that of the magnetic moments of the surrounding Cr atoms [8].

The neutron diffraction patterns at 300 K show a single peak around both Cr(001) and Cr(010) for all the multilayers with the Cr layer thickness from 40 to 160 Å. The diffraction patterns for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ multilayers ($t_{\mathrm{Cr}} = 40$, 120, and 160 Å) around Cr(010) along the L axis are shown in Fig. 1(a). The abscissa is expressed by the reciprocal lattice unit (r.l.u.) of $2\pi/a_{001}$, where a_{001} is the average lattice parameter in the growth direction. The result indicates that, at room temperature, the Cr in all theses samples has a commensurate SDW with a simple antiferromagnetic (CAF) structure, where the magnetic moments align ferromagnetically in one (001) plane and antiferromagnetically between adjacent (001) planes. The CAF structure is kept in phase across the Sn layer, with the magnetization of atomic (001) planes on both sides of the monatomic Sn layer being parallel with each other. The width of the fundamental peak reaches the resolution limit, indicating that the magnetic correlation is well established across the inserted Sn layers, although the satellite peaks due to the superstructure do not appear clearly. The stabilization of a CAF phase around room

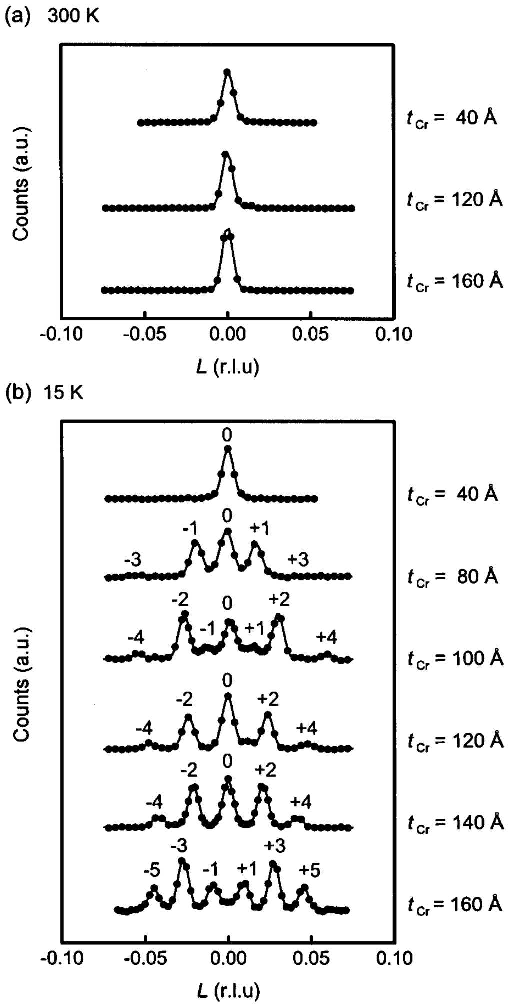

FIG. 1. Neutron diffraction patterns around Cr(010) along the L direction for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ at (a) 300 K and (b) around 15 K. The peak positions for the patterns at 15 K are identified with a unit of $\Delta \equiv a_{001}/(2\Lambda_{\mathrm{SL}})$ as L (r.l.u.) $= \pm N\Delta$ ($N = 0, 1, 2, \ldots$) (see the text).

temperature, with a much higher magnetic transition temperature than the bulk, has also been reported for pure Cr films [9].

The ^{119}Sn Mössbauer spectra for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ multilayers ($t_{\mathrm{Cr}} = 40$ and 80 Å) at 300 K are shown in Fig. 2(a). A magnetically split six-line component with a large hyperfine field is observed in the spectra. The hyperfine field has a Gaussian-like distribution with the maximum around 10 or 11 T and no component around 0 T. This is in contrast with the spectra of dilutely dissolved Sn in bulk Cr, where the hyperfine field is distributed continuously, typically, from 8 to 0 T [10]. The above features in the spectra are common for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ multilayers although the hyperfine field (the peak value) decreases monotonously from 11 to 9 T when t_{Cr} increases from 40 to 160 Å. As indicated by the neutron diffraction patterns, the Cr has a CAF structure, and the coherency of the CAF structure is kept across the inserted Sn layer. The

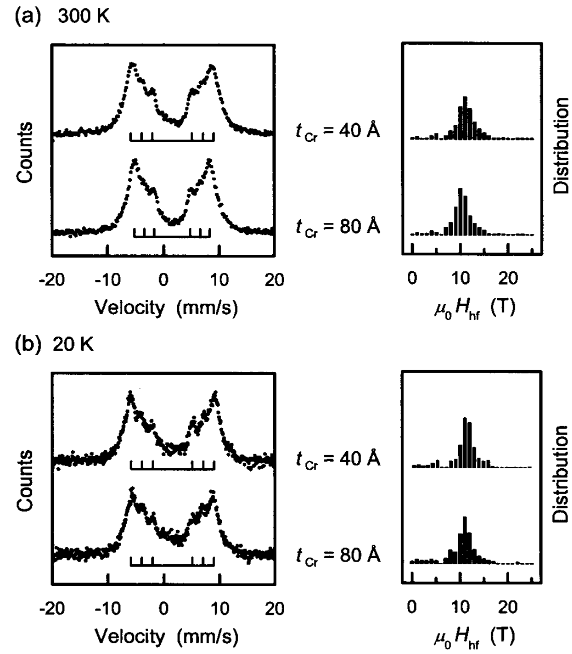

FIG. 2. ^{119}Sn Mössbauer spectra for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ multilayers ($t_{\mathrm{Cr}} = 40$ and 80 Å) at (a) 300 K and (b) 20 K. The distribution of the magnetic hyperfine field obtained from the fitting of the spectra is shown on the right.

appearance of a finite hyperfine field at the Sn sites also shows that the magnetic moments of the atomic Cr(001) planes that are sandwiching the Sn layer are oriented to the same direction; if the magnetic moments of atomic Cr(001) planes in both sides of the Sn layer were oriented to the opposite direction with each other, the magnetic hyperfine field transferred at the Sn site would be canceled to zero [11]. Thus, the substitution of a monatomic Sn layer for an atomic Cr(001) plane does not disturb the coherency of the antiferromagnetic order in Cr. The observed large hyperfine field suggests that the magnetic moments of Cr around Sn are somewhat enhanced [11].

When the samples are cooled down to low temperatures, the neutron diffraction patterns change drastically. The patterns around Cr(010) along the L direction for $[\mathrm{Cr}(t_{\mathrm{Cr}})/\mathrm{Sn}(2\ \text{Å})]$ multilayers ($t_{\mathrm{Cr}} = 40, 80, 100, 120, 140$, and 160 Å) at around 15 K are shown in Fig. 1(b). The patterns show large dependence on the Cr layer thickness t_{Cr} or the superlattice period Λ_{SL} ($= t_{\mathrm{Cr}} + 2$ Å). When t_{Cr} is equal to 40 Å, the pattern has a single peak, indicating that the CAF structure persists down to low temperatures. For the samples with $t_{\mathrm{Cr}} \geq 80$ Å, several satellite peaks appear, suggesting that SDW with the modulation along the film normal appears at low temperatures. All the peak positions for all the samples can be indexed by a unit of $\Delta \equiv a_{001}/(2\Lambda_{\mathrm{SL}})$ as $\pm N\Delta$ (r.l.u.) or by a unit of $\delta Q \equiv 2\pi/(2\Lambda_{\mathrm{SL}})$ as $\pm N\delta Q$ (Å^{-1}) ($N = 0, 1, 2, \ldots$). This means that the wavelength of the Fourier component of the SDW modulation, Λ_{mod}, can be expressed as $\Lambda_{\mathrm{mod}} = 2\Lambda_{\mathrm{SL}}/N$ for each sample. Thus, the neutron patterns for the Cr/Sn multilayers are quite different from those for pure bulk Cr, where the SDW modulation is composed of an almost pure sinusoidal function [1,12].

The positions of the neutron diffraction peaks around Cr(001) along the L direction are also identified as $\pm N\Delta$, indicating the same modulation length as obtained from the patterns around Cr(010). Modulation along the in-plane K direction was not observed in the K scans around either Cr(001) or Cr(010) for any samples in Fig. 1(b). The detail of the scans for these axes will be published elsewhere [13].

Although the neutron patterns change drastically at low temperatures, the Mössbauer spectra do not change much when the temperature decreases to 20 K [Fig. 2(b)]. The basic feature of the distribution in hyperfine fields remains unchanged, except that the hyperfine field (the peak value) increases by about 1 T. This means that the local magnetic structure around the Cr/Sn interfaces has no significant change between 300 and 20 K, except that the size of the enhanced magnetic moments increases a little as the temperature decreases.

With the results from both neutron diffraction and Mössbauer spectroscopic measurements taken into consideration, it can be concluded that the enhanced magnetic moments of Cr at the Cr/Sn interfaces pin the antinode of the SDW modulation which appears at low temperatures. On the basis of this idea, the neutron results in Fig. 1(b) are interpreted in the following way. As already mentioned, the neutron diffraction peaks at L (r.l.u.) $= \pm N\Delta$ are attributed to the Fourier components of SDW modulation with the wavelength $\Lambda_{\mathrm{mod}} = 2\Lambda_{\mathrm{SL}}/N$. Each sinusoidal Fourier component is illustrated in Fig. 3(a) in such a way that antinodes are pinned at the Sn layers. The integer N corresponds to the numbers of nodes in a Cr layer in this case. For the samples with $t_{\mathrm{Cr}} =$ 120, and 140 Å, the peaks at $L = \pm 2\Delta$ are dominant (except the peak at $L = 0$), so that the main modulation has antinodes at the interfaces and two nodes in the Cr layer. The coexistence of the peak at $L = 0$ indicates that the SDW modulation has a constant Fourier component and is not composed of a pure sinusoidal function. Because of the enhancement of the magnetic moments at the interfaces, it is expected that the modulation amplitude is larger for the antinodes at the interfaces and smaller for the antinode at the center of the Cr layer [Fig. 3(b)]. The distorted SDW modulation also has a higher order modulation, which is the origin of the peaks at $L = \pm 4\Delta$, although the phase of the component cannot be determined from the neutron results. (Note here that we cannot exclude a possibility that a part of the peak at $L = 0$ is from another independent CAF phase.) For the sample with $t_{\mathrm{Cr}} = 160$ Å, the main modulation is expressed by the Fourier component with $N = 3$, which has three nodes in the Cr layer. The enhancement of magnetic moments at the interfaces [Fig. 3(b)] and accompanying

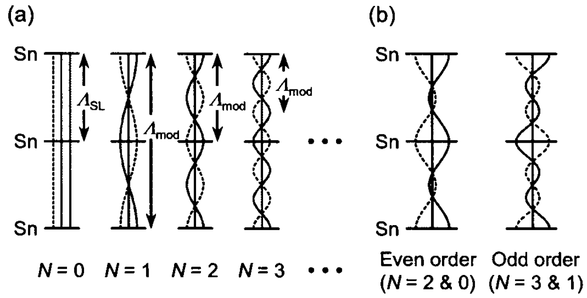

FIG. 3. (a) Illustration of Fourier components of magnetic modulation in real space, which cause neutron diffraction peaks at L (r.l.u.) $= \pm N\Delta$. It is assumed for each component that antinodes are pinned at the Sn layers. (b) Distorted SDW modulation, which can be expressed by the sum of the even-order Fourier components (e.g., $N = 2$ plus $N = 0$) or the odd-order Fourier components (e.g., $N = 3$ plus $N = 1$).

distortion in the SDW modulation cause the Fourier components with $N = 1$ and 5. It should be noted that the samples with $t_{\mathrm{Cr}} = 120$ and 140 Å have only even-order Fourier components and the sample with $t_{\mathrm{Cr}} = 160$ Å has odd-order components. This fact supports the interpretation that the different satellite peaks for each sample are from a single distorted SDW phase, and not from several independent pure sinusoidal SDW phases. The wavelength of the main Fourier component of the SDW modulation, which is obtained from the neutron diffraction patterns in Fig. 1(b), is plotted in Fig. 4 as a function of the superlattice period. As clearly demonstrated, the main Fourier component Λ_{SDW} changes discretely as a function of Λ_{SL}. A distorted SDW structure with an antinode at the center of the Cr layer can be reproduced only from even-order Fourier components, and that with a node at the center of the Cr layer can be reproduced only from odd-order components [Fig. 3(b)]. In the neutron diffraction patterns for the samples with $t_{\mathrm{Cr}} = 100$ and 80 Å, both odd- and even-order peaks are observed. As shown in Fig. 4, these samples are near the thickness boundary between two different SDW phases with a different number of nodes. Therefore, it is thought that the odd- and even-order peaks are from two different phases, which may be caused by an inevitable imperfection in the multilayered structure.

Now we briefly comment on the direction of magnetic moments in the Cr layers. From the intensity ratio of the magnetically split sextet in the Mössbauer spectra, which is sensitive to the direction of the magnetic hyperfine field, it is obvious that the magnetic moments are not perfectly along the film plane or the film normal. If it is assumed that the axes of easy magnetization in the Cr/Sn multilayers are along the same crystallographic axes as those in bulk Cr, it is expected that a multidomain structure with the direction of magnetic moments along the [100], [010], and [001] directions is formed in the Cr layers. The Mössbauer results suggest that the ratio of

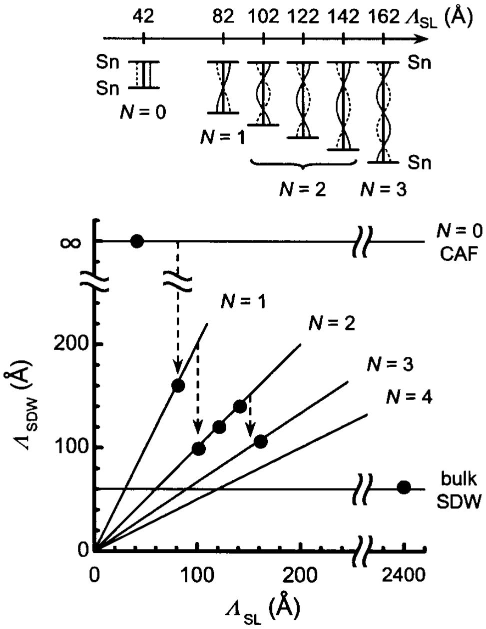

FIG. 4. Wavelength of the main Fourier component of SDW modulation (Λ_{SDW}) obtained from the neutron diffraction patterns around 15 K as a function of the artificial superlattice period (Λ_{SL}). The change of modulation as a function of Λ_{SL} is shown also in real space. Only the main Fourier component is illustrated for simplicity, although the SDW modulation in real multilayers is distorted from a pure sinusoidal function.

the domains with in-plane magnetic moments to those with perpendicular magnetic moments is about 2:3. For the neutron diffraction measurements, the scans around Cr(010) are sensitive to both in-plane and out-of-plane components and those around Cr(001) are sensitive only to the in-plane components. A rough comparison of the peak intensity around Cr(010) and that around Cr(001) and a preliminary result from polarized neutron diffraction measurement suggest that the magnetic moments are partially in the film plane and partially along the film normal. We stress here that the important thing is that for both in-plane and out-of-plane spin components the SDW is modulated along the growth direction and the wavelength is harmonic with the superlattice period.

It is expected theoretically that the magnetic moments of Cr are enhanced at a vacuum interface [1]. In the present system, the inserted Sn layer gives a similar effect to "vacuum" for the $3d$ electrons of Cr, resulting in an enhancement of magnetic moments at the interface [8], whereas the spin polarization of $5s$ electrons in the Sn layer mediates the coherency of the antiferromagnetic structure across the Sn layer. Distorted SDW structures with antinodes pinned at the interfaces are predicted from first principles calculations for Fe/Cr layered structures with ideally flat interfaces [14,15]. An oscillatory change in the modulation length of SDW and an accompanying discrete change in the number of nodes in a Cr layer as a function of the Cr layer thickness are also predicted for Fe/Cr/Fe trilayers with ideally flat interfaces [16]. Indirect experimental evidence to support this prediction has been shown by scanning electron microscopy with polarization analysis [3,4]. The results from the neutron diffraction experiments on Fe/Cr multilayers, on the other hand, indicate that the wavelength of the SDW modulation is not much dependent on the Cr layer thickness in real multilayers, which have rough interfaces and an accompanying magnetic frustration effect [17]. The system in the present work has no magnetic frustration effect even if the Sn layers are not ideally flat, since the Sn atoms are substituted for Cr atoms without disturbing the antiferromagnetic coherency in the Cr film. As a result of this, the discrete change in the SDW modulation is well demonstrated as shown above.

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